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4f spin dynamics in $TbNi_2B_2C$ by ^{11}B NMR

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Abstract

^{11}B NMR measurements have been performed to investigate local electronic structure and 4f spin dynamics for $TbNi_2B_2C$ single crystal. ^{11}B NMR spectra show three resonance peaks due to the quadrupolar interaction. Shift and linewidth are huge and strongly temperature-dependent. In addition, both are proportional to magnetic susceptibility, indicating that the hyperfine field at the boron site originates from the 4f spins of Tb. ^{11}B NMR shift and relaxation rates show high anisotropy for field parallel and perpendicular to the c-axis. Anisotropy of the shift and the relaxation rates suggests that the hyperfine field perpendicular to the c-axis is larger.

Keywords : $TbNi_2B_2C$, 4f moment, ^{11}B NMR, shift and relaxation rates

I. Introduction

RNi_2B_2C (R=rare-earth) compounds provide a unique chance to study the interplay between superconductivity and long-range magnetic order. The grounds state of these compounds range from superconductivity (R=Lu, Sc, Y, Th), to superconductivity coexisting with magnetic order (R=Tm, Er, Ho, Dy), and to magnetic order without superconductivity (R=Gd, Tb). [1-7] The layered structure [8] of these compounds, which consists of R-C layers well separated from the conducting Ni_2-B_2 layers, resembles that of the cuprate superconductors. However, the band structure calculations [9] indicate these systems are rather close to conventional superconductors. The layered structure isolates the conduction electron from the magnetic R ions and consequently generates competition between superconductivity and magnetic ordering.

It has been reported that $TbNi_2B_2C$ does not show any indication of superconductivity down to 2 K [2]. The neutron diffraction study [10] confirms that this compound exhibits an antiferromagnetic order below 15 K. Also magnetic properties [11] and transport measurements [12] show that magnetic properties are highly anisotropic and magnetic structures in the ordered state are rather complex. In this paper, we present results of ^{11}B NMR measurements to investigate the 4f spin dynamics of Tb ion as well as local electronic and magnetic structures.

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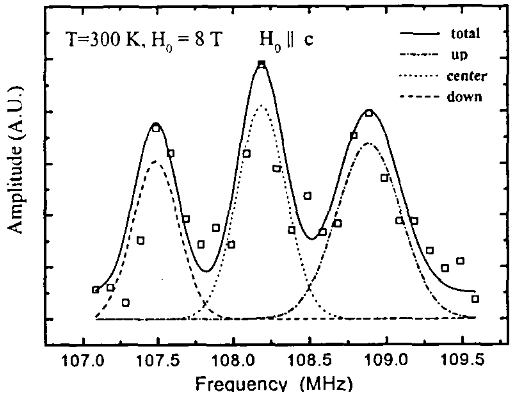
II. Experiments

$TbNi_2B_2C$ single-crystal specimens were grown by the high-temperature Ni_2B flux technique as described elsewhere [13]. The sample size was roughly 8x8x2 mm^3. The pulsed ^{11}B NMR measurements were carried out at 4.7 and 8.0 T parallel and perpendicular to the c-axis at various temperatures. The measurements at 4.7 T were carried out at Seoul Branch of Korea Basic Science Institute. The phase-alternating pulse sequences were employed to significantly reduce the electro-mechanical vibration (ring-down) after pulses [14]. The broad spectra were scanned by the point-by-point method changing a spectrometer frequency. The cryogenic measurements were performed in the Oxford continuous flow cryostat (CF1200N).

III. Results and Discussion

Figure 1 shows ^{11}B NMR spectrum of $TbNi_2B_2C$ at 300 K for field parallel to the c-axis. The spectrum shows three resonance peaks, one central (1/2<->-1/2) and two satellite (3/2<->1/2 and -1/2<->-3/2) transitions. These originate from the quadrupolar splitting of nuclear Zeeman levels for ^{11}B nucleus of I=3/2. The separation between the central and the satellite peaks is the nuclear quadrupole frequency, ν_Q, which is determined by the electric quadrupole moment of ^{11}B nucleus, Q, and the electric field gradient (EFG), V_{zz}, at the boron site. From variation of the separation for magnetic field direction relative to the c-axis, we find that the c-axis is the principal axis of EFG tensor at the boron sites.



Linewidths of the three peaks are much broader than those of a nonmagnetic homologue YNi_2B_2C [15]. Thus it is clear that the broad linewidths originate from magnetic broadening due to the 4f moments of Tb.

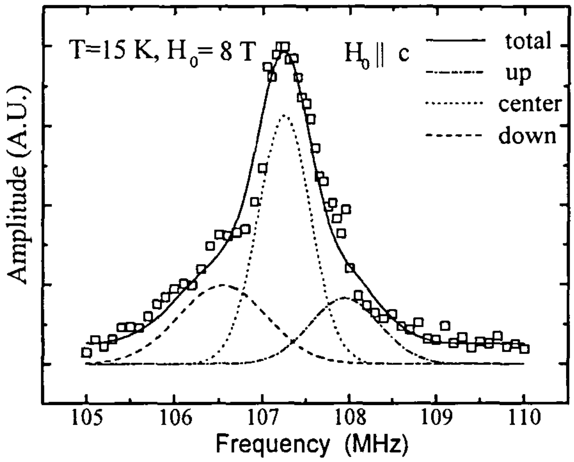
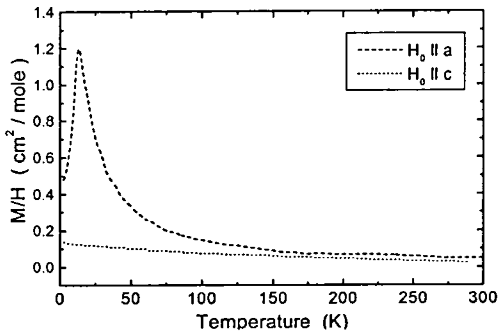


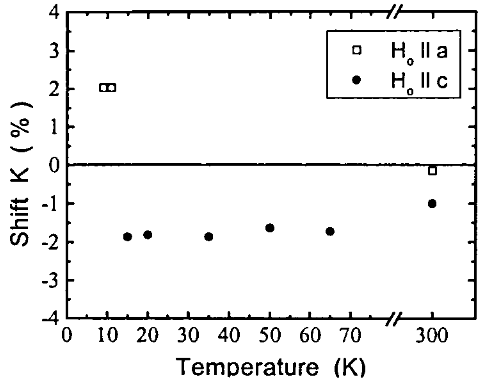
Figure 2 shows ^{11}B NMR spectrum at 15 K for field parallel to the c-axis. The three resonances become much broader giving rise to a single broad peak. This magnetic broadening comes from the larger susceptibility of $TbNi_2B_2C$ at low temperature. The susceptibility data are shown in Figure 3. Also, the center of the peak is shifted to a low frequency side, which means that the shift is negative for field parallel to the c-axis. Both the shift and the linewidth increase at low temperatures.

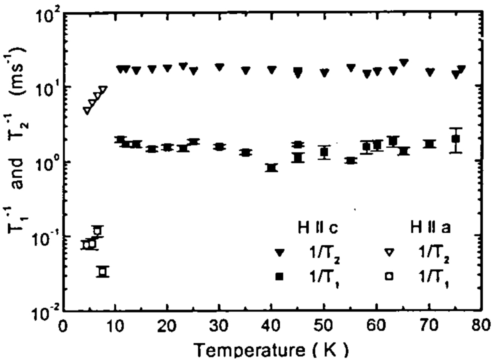
^{11}B NMR shifts for field parallel and perpendicular to the c-axis are plotted in Figure 4. The shift is positive for field perpendicular to the c-axis, suggesting that major hyperfine fields at the boron nuclei are from the 2s orbital of boron. On the other hand, the shift is negative for field parallel to the c-axis, indicating that the total hyperfine field at the boron nuclei is from core-polarization of boron orbitals by 4f moments. The shift is huge, strongly temperature-dependent, and proportional to the magnetic susceptibility. This behavior confirms that the shift has a major contribution from 4f moment



polarization. In order to check this idea, we have to plot the shift versus the susceptibility using temperature as a parameter. Then the plot is going to show a straight line suggesting that the shift scales with the susceptibility. The slope of the straight line is a hyperfine coupling constant A; that is, $A=K\chi$. In doing so, we can obtain anisotropy of the hyperfine coupling constant A. Linewidths also increase at low temperature. The linewidths for three resonance peaks scale with the susceptibility. Therefore the origin of the linewidths is magnetic broadening due to the 4f moments of Tb ions.

Information regarding dynamics of 4f spins can be found from relaxation rates of NMR. The spin-lattice relaxation rate, $1/T_1$, and the spin-spin relaxation rate, $1/T_2$, are plotted in Figure. 5. $1/T_1$ and $1/T_2$ are identical for field parallel and perpendicular to the c-axis. At high temperature, both relaxation rates are weakly temperature-dependent, which is a typical behavior for random spin fluctuation. Generally speaking, however, both relaxation rates tend to increase if local spins slow down as temperature decreases toward a magnetic ordering temperature. At lower temperature, both relaxation rates start to be much different depending on the magnetic field direction and become highly anisotropic suggesting that there is a significant change in the 4f spin dynamics.





Both $1/T_1$ and $1/T_2$ probe local field fluctuation. Difference between two relaxation rates is direction and time scale of fluctuating local field. Namely, $1/T_1$ sensitively probes the component of fluctuating local field perpendicular to the external field at the NMR frequency scale. On the other hand, $1/T_2$, in addition to the same contribution of $1/T_1$, is also sensitive to the component of slowly fluctuating local field parallel to the external field at the frequency scale much smaller than the NMR frequency.

Therefore, the anisotropy of $1/T_1$ and $1/T_2$ can provide information about microscopic details of spin dynamics. In Figure 5, both relaxation rates are larger for field parallel to the c-axis. This fact indicates the in-plane component of fluctuating local field due to 4f spins is larger. It should be noticed that the *static* in-plane hyperfine field is also larger based on the fact that the in-plane shift is larger, as shown in Figure 4.

IV. Summary

^{11}B pulsed NMR measurements have been performed to investigate local electronic structure and 4f spin dynamics for $TbNi_2B_2C$ single crystal. ^{11}B NMR spectra show three resonance peaks due to the quadrupolar interaction. ^{11}B NMR shift and linewidth are huge and strongly temperature-dependent. In addition, both are proportional to magnetic susceptibility, indicating that the hyperfine field at the boron site originates from the 4f spins of Tb. The linewidths are dominated by magnetic broadening due to the 4f moments. The shift and the spin-lattice relaxation rate show a high anisotropy for field parallel and perpendicular to the c-axis. Anisotropy of the shift and the relaxation rates suggests that the hyperfine field perpendicular to the c-axis is larger.

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